Direct Aerosol Absorption Measurements using Photothermal Interferometry

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Introduction

One of the goals of the atmospherics community is to better quantify the role of aerosol absorption on the radiative balance of the atmosphere. Black carbon (BC) is important since it absorbs light thereby contributing to global warming. However, the level of uncertainty regarding the contribution of aerosols (BC & BC-containing particles) to radiative forcing is poorly understood.

The goal of this project is to develop a method for directly measuring aerosol absorption that is capable of aircraft deployment.

Photothermal Methods for Measuring Aerosol Absorption

Photothermal Spectroscopy (PTS), which relies on the thermal dissipation of spectrally absorbed energy for its signal, offers the highly desirable trait of directly measuring aerosol absorption

Two PTS –based approaches are currently under development or in use for aerosol absorption:

Photoacoustic Spectroscopy *(Arnott & co-workers, 1998, 1999, 2005; Lack, et al., 2006)*

&

Photothermal Interferometry *(Sedlacek, 2006)*

The hallmark of any photothermal-based technique is that it <u>directly</u> measures aerosol absorption <u>without interference from aerosol scattering</u>

Photothermal Interferometry

Interferometry is the study of the interference patterns created by combining two sets of coherent light waves that have a constant phase relation and same polarization. This very sensitive technique can be used to follow the thermal dissipation that characterizes PTS.

$$E_r = A_r e^{i(\omega_r + \phi_r)} \qquad E_p = A_p e^{i(\omega_p + \phi_p + \Delta\phi(t))}$$

where $\Delta\phi(t) = m\sin(\omega_{mod} t)$

Intensity = $(E_r + E_p)(E_r + E_p)^* \ldots$

$$I(t) = \{A_p^2 + A_r^2 + 2A_pA_r[\cos(\phi_r - \phi_p)\cos\Delta\phi(t) - \sin(\phi_r - \phi_p)\sin\Delta\phi(t)]\}$$

In the limit of $\Delta\phi << 1$; $\phi_r - \phi_p = \pi/2$

$$I(t) \propto C\Delta\phi(t)$$

Photothermal Interferometry: Performance Estimate

$\Delta\phi_{min}$ = 1.6 x 10^{-8} rad
r = 0.5 mm
ρ = 1.205 kg m^{-3}
C_p = 1005.4 J kg^{-1} K^{-1}
ω_{mod} = 100 Hz
λ = 632.8 nm
l = 5 cm
n = 1.000292
P_{exc} = 250 mW
T_o = 293 K

$$\alpha_{min} = \frac{\Delta\phi_{min}\, 2r^2 \rho C_p \omega_{mod} \lambda T_o}{\pi l (n-1) P_{exc}}$$

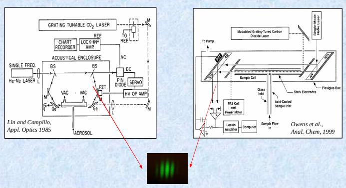
α_{min} ~ 5 x 10^{-8} m^{-1} = 0.05 Mm^{-1} = 50 Gm^{-1}

Relevant PTI Studies

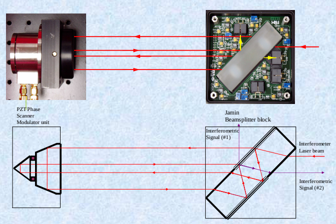
Lin and Campillo (1985) used PTI to measure $(NH_4)_2SO_4$ *absorption using a Mach-Zehnder interferometer*

Detection sensitivity of 50 Mm^{-1} (λ_{abs}=1086 cm^{-1}) *– sensitivity was limited by bkgnd water vapor absorption at this wavelength*

More recently Davis and co-workers (1999) using a modified Jamin interferometer reported a sensitivity of 37 Gm^{-1} *for* NH_3 (λ_{abs}=1085 cm^{-1}).



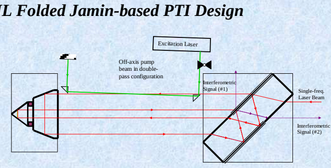
Folded Jamin Interferometer Design offers superior optical stability



- *Insensitive to rotation and tilt*
- *Simple positional alignment*
- *Common path enables common mode rejection providing increased robustness, stability (mechanical vibrations) and sensitivity*
- *Compact size*
- *Maximal pathlength for given size (inherent double-pass configuration)*

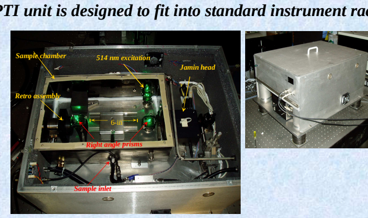
AIMS folded Jamin interferometer manufactured by Interferomet Inc.: http://www.interferomet.com

BNL Folded Jamin-based PTI Design

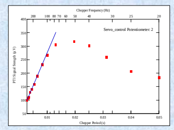


Glancing configuration eliminates all spurious signals

PTI unit is designed to fit into standard instrument rack

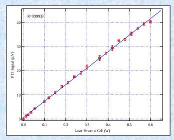


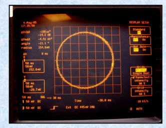
Subsystem Performance Characteristics



Servo-control requirements: Maintain system at quadrature by correcting for slowing varying drifts (e.g., thermal) but passing high-frequency signal onto LIA.

Drifts on the order to <100 Hz are corrected by feedback circuit



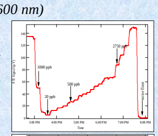


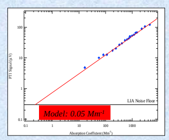
Use Lissajous circle to optimize unit

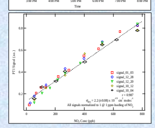
Unit Calibration Measurements: NO_2 *@ 514 nm*

Calibration standard (410 - ~600 nm)

Relevant PSD parameters:
Detection bandwidth: 7.8 mHz
Pathlength: 5 cm
RMS noise floor: 300 nV







Measured absorption cross-section compares favorably with previous literature values:

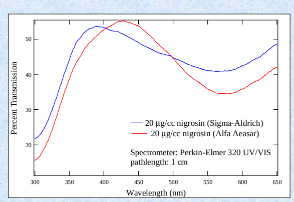
$$\sigma_{abs}^{pti} = 2.2 \pm 0.08 \times 10^{-19} cm^2 molec^{-1}$$

$$\sigma_{abs}^{lit} = 1.9 \pm 0.18 \times 10^{-19} cm^2 molec^{-1}$$ *(Average of 4 different studies*)*

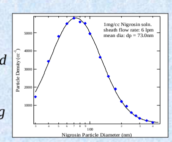
**Vandaele et al., JAC, 25 1996; Davidson et al., JGR, 93, 1988; Harder et al., JGR, 103, 1997; Vandaele et al., JQSRT, 59, 1998*

Evaluation of PTI unit Towards Measurement of Aerosol Absorption: Nigrosin

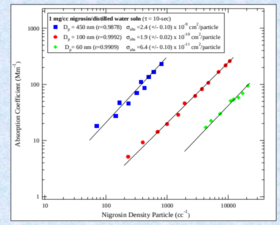
Nigrosin is a polyaniline-based black pigment. Its broad, featureless absorption spectrum in the visible makes it an ideal BC surrogate for instrument evaluation.



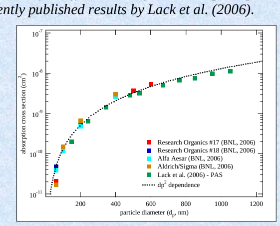
Size-selected nigrosin aerosols were generated using TSI constant output atomizer and DMA and counted using CPC.



Using our calibration standard (NO_2)*, the absorption coefficient* (Mm^{-1}) *of nigrosin aerosols as a function of particle diameter can be determined. Representative data for three different particle diameters is shown below.*

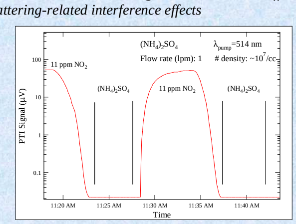


Measurement of the PTI signal as a function of nigrosin number density gives a slope that is equal to the absorption cross-section (cm^2*/particle). Shown below are some measurements of* σ_{abs} *for various nigrosin particle diameters for different manufacturers superimposed on the recently published results by Lack et al. (2006).*



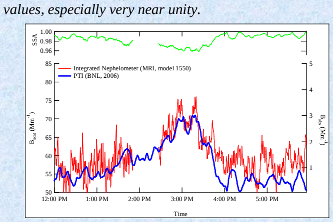
Measurements on non-absorbing aerosols: $(NH_4)_2SO_4$

Despite the concentration being <u>4-5 orders-of-magnitude</u> higher than that used in the nigrosin experiments, no signal was observed, demonstrating that PTI does not suffer from scattering-related interference effects



Measurement of Ambient Aerosols

Experiments have recently been initiated to measure ambient aerosols at BNL using PTI (B_{abs}*) and an integrating nephelometer (*B_{scat}*). The ability of the PTI technique to directly measure* B_{abs} *enable accurate measurement of SSA values, especially very near unity.*



Publications

Sedlacek, A. J., "Real-time detection of ambient aerosols using photothermal interferometry: Folded Jamin interferometer" Rev. Sci. Instr. ***77*** *064903 (2006)*

Acknowledgements

Authors would like to thank Dr. J. Wang for use of his DMA and CPC .